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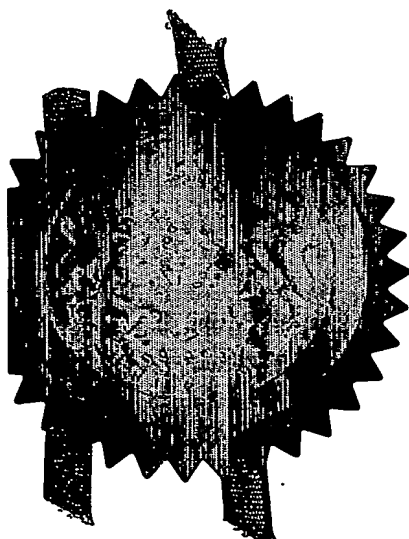
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Signed

*Evans*

Dated

10 February 2003

Patent Act 1977  
(Rule 16)

6 MAR 2002



06HAR02-5704110-1 C12133  
P01/7700 0.00-0205212.4

# Request for grant of a patent

(See the notes on the back of this form. You can also get an explanatory leaflet from the Patent Office to help you fill in this form)

The Patent Office

Cardiff Road  
Newport  
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1. Your reference

473

2. Patent application number

(The Patent Office will fill in this part)

0205212.4

3. Full name, address and postcode of the or of each applicant (underline all surnames)

ELAM-T LIMITED  
103 BOROUGH ROAD  
LONDON  
SE1 OAA  
GB

Patents ADP number (if you know it)

If the applicant is a corporate body, give the country/state of its incorporation

8105280001

4. Title of the invention

ELECTROLUMINESCENT MATERIALS

5. Name of your agent (if you have one)

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

COHEN, ALAN NICOL  
2 GROVE PLACE  
TATSFIELD  
Nr. WESTERHAM  
KENT  
TN16 2BB

Patents ADP number (if you know it)

6963557001

6. If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and (if you know it) the or each application number

Country

Priority application number  
(if you know it)

Date of filing  
(day / month / year)

7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application

Date of filing  
(day / month / year)

8. Is a statement of inventorship and of right to grant of a patent required in support of this request? (Answer 'Yes' if:

YES

a) any applicant named in part 3 is not an inventor, or

b) there is an inventor who is not named as an applicant, or

c) any named applicant is a corporate body.

See note (d)).

**Patents Form 1/77**

9. Enter the number of sheets for any of the following items you are filing with this form.  
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Continuation sheets of this form

Description 30

Claim(s) 6

Abstract 1

Drawing(s) 15 x 16

10. If you are also filing any of the following, state how many against each item.

Priority documents

Translations of priority documents

Statement of inventorship and right to grant of a patent (*Patents Form 7/77*)

Request for preliminary examination and search (*Patents Form 9/77*)

Request for substantive examination (*Patents Form 10/77*)

Any other documents  
(please specify)

11.

I/We request the grant of a patent on the basis of this application.

Signature



Date

01/03/02

12. Name and daytime telephone number of person to contact in the United Kingdom

A. N. Cohen

01959 577172

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## Electroluminescent Device

The present invention relates to electroluminescent devices and displays.

5 Materials which emit light when an electric current is passed through them are well known and used in a wide range of display applications. Liquid crystal devices and devices which are based on inorganic semiconductor systems are widely used, however these suffer from the disadvantages of high energy consumption, high cost of manufacture, low quantum efficiency and the inability to make flat panel displays.

10

With organic light emitting polymers it is not possible to obtain pure colours, they are expensive to make and have a relatively low efficiency.

15

Patent application WO98/58037 describes a range of lanthanide complexes which can be used in electroluminescent devices which have improved properties and give better results. Patent Applications PCT/GB98/01773, PCT/GB99/03619, PCT/GB99/04030, PCT/GB99/04024, PCT/GB99/04028, PCT/GB00/00268 describe electroluminescent complexes, structures and devices using rare earth chelates.

20

In order to modify the light emitted a fluorescent dye can be incorporated in the electroluminescent layer or in the electron transmitting layer.

25

According to the invention there is provided an electroluminescent device comprising in sequence, an anode, a layer of an electroluminescent material of general formula  $(La)_nM$  where M is a rare earth, lanthanide or an actinide, La is an organic complex and n is the valence state of M and a cathode, in which the layer of an electroluminescent material includes a fluorescent dye.

The electroluminescent compounds which can be used in the present invention are of general formula  $(L\alpha)_nM$  where M is a rare earth, lanthanide or an actinide,  $L\alpha$  is an organic complex and n is the valence state of M.

- 5 Other electroluminescent compounds which can be used in the present invention are of formula



10

where  $L\alpha$  and  $L_p$  are organic ligands, M is a rare earth, transition metal, lanthanide or an actinide and n is the valence state of the metal M. The ligands  $L\alpha$  can be the same or different and there can be a plurality of ligands  $L_p$  which can be the same or different.

15

For example  $(L_1)(L_2)(L_3)(L_{..})M (L_p)$  where M is a rare earth, transition metal, lanthanide or an actinide and  $(L_1)(L_2)(L_3)(L_{..})$  are the same or different organic complexes and  $(L_p)$  is a neutral ligand. The total charge of the ligands  $(L_1)(L_2)(L_3)(L_{..})$  is equal to the valence state of the metal M. Where there are 3 groups  $L\alpha$  which corresponds to the III valence state of M the complex has the formula  $(L_1)(L_2)(L_3)M (L_p)$  and the different groups  $(L_1)(L_2)(L_3)$  may be the same or different

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$L_p$  can be monodentate, bidentate or polydentate and there can be one or more ligands  $L_p$ .

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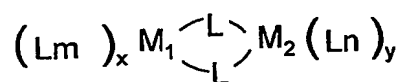
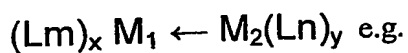
Preferably M is metal ion having an unfilled inner shell and the preferred metals are selected from Sm(III), Eu(II), Eu(III), Tb(III), Dy(III), Yb(III), Lu(III), Gd (III), Gd(III) U(III), Tm(III), Ce (III), Pr(III), Nd(III), Pm(III), Dy(III), Ho(III), Er(III), Yb(III) and more preferably Eu(III), Tb(III), Dy(III), Gd (III).

30

Further electroluminescent compounds which can be used in the present invention are of general formula  $(L\alpha)_n M_1 M_2$  where  $M_1$  is the same as M above,  $M_2$  is a non rare earth metal,  $L\alpha$  is as above and  $n$  is the combined valence state of  $M_1$  and  $M_2$ . The complex can also comprise one or more neutral ligands  $Lp$  so the complex has the general formula  $(L\alpha)_n M_1 M_2 (Lp)$ , where  $Lp$  is as above. The metal  $M_2$  can be any metal which is not a rare earth, transition metal, lanthanide or an actinide examples of metals which can be used include lithium, sodium, potassium, rubidium, caesium, beryllium, magnesium, calcium, strontium, barium, copper (I), copper (II), silver, gold, zinc, cadmium, boron, aluminium, gallium, indium, germanium, tin (II), tin (IV), antimony (II), antimony (IV), lead (II), lead (IV) and metals of the first, second and third groups of transition metals in different valence states e.g. manganese, iron, ruthenium, osmium, cobalt, nickel, palladium(II), palladium(IV), platinum(II), platinum(IV), cadmium, chromium, titanium, vanadium, zirconium, tantalum, molybdenum, rhodium, iridium, titanium, niobium, scandium, yttrium.

For example  $(L_1)(L_2)(L_3)(L...)M (Lp)$  where  $M$  is a rare earth, transition metal, lanthanide or an actinide and  $(L_1)(L_2)(L_3)(L...)$  and  $(Lp)$  are the same or different organic complexes.

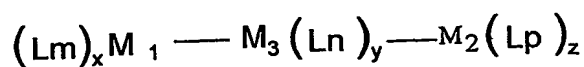
Further organometallic complexes which can be used in the present invention are binuclear, trinuclear and polynuclear organometallic complexes e.g. of formula



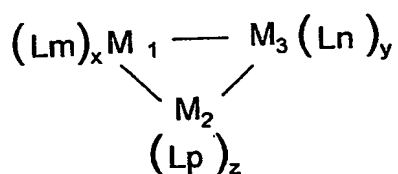
where  $L$  is a bridging ligand and where  $M_1$  is a rare earth metal and  $M_2$  is  $M_1$  or a non rare earth metal,  $Lm$  and  $Ln$  are the same or different organic ligands  $L\alpha$  as defined above,  $x$  is the valence state of  $M_1$  and  $y$  is the valence state of  $M_2$ .

In these complexes there can be a metal to metal bond or there can be one or more bridging ligands between  $M_1$  and  $M_2$  and the groups  $Lm$  and  $Ln$  can be the same or different.

- 5 By trinuclear is meant there are three rare earth metals joined by a metal to metal bond i.e. of formula



- 10 or

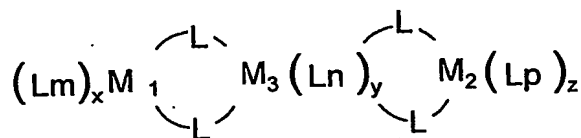


- 15 where  $M_1$ ,  $M_2$  and  $M_3$  are the same or different rare earth metals and  $Lm$ ,  $Ln$  and  $Lp$  are organic ligands  $Lx$  and  $x$  is the valence state of  $M_1$ ,  $y$  is the valence state of  $M_2$  and  $z$  is the valence state of  $M_3$ .  $Lp$  can be the same as  $Lm$  and  $Ln$  or different.

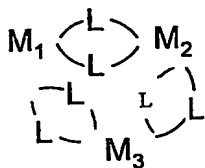
The rare earth metals and the non rare earth metals can be joined together by a metal to metal bond and/or via an intermediate bridging atom, ligand or molecular group.

- 20

For example the metals can be linked by bridging ligands e.g.

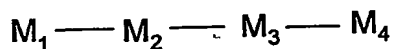


- 25 or



where L is a bridging ligand

- 5 By polynuclear is meant there are more than three metals joined by metal to metal bonds and/or via intermediate ligands

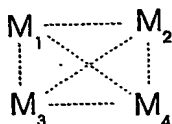


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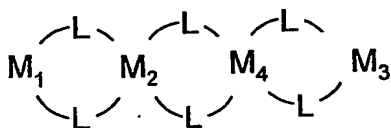
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or



- 15 or

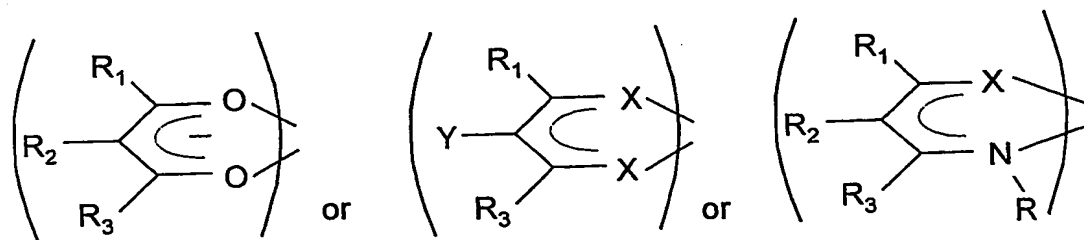


where  $M_1$ ,  $M_2$ ,  $M_3$  and  $M_4$  are rare earth metals and L is a bridging ligand.

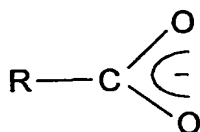
Preferably  $L\alpha$  is selected from  $\beta$  diketones such as those of formulae

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- where  $R_1$ ,  $R_2$  and  $R_3$  can be the same or different and are selected from hydrogen, and substituted and unsubstituted hydrocarbyl groups such as substituted and unsubstituted aliphatic groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoromethyl groups, halogens such as fluorine or thiophenyl groups;  $R_1$ ,  $R_2$  and  $R_3$  can also form substituted and unsubstituted fused aromatic, heterocyclic and polycyclic ring structures and can be copolymerisable with a monomer e.g. styrene. X is Se, S or O,
- Y can be hydrogen, substituted or unsubstituted hydrocarbyl groups, such as substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorine, fluorocarbons such as trifluoromethyl groups, halogens such as fluorine or thiophenyl groups or nitrile.
- Examples of  $R_1$  and/or  $R_2$  and/or  $R_3$  include aliphatic, aromatic and heterocyclic alkoxy, aryloxy and carboxy groups, substituted and unsubstituted phenyl, fluorophenyl, biphenyl, phenanthrene, anthracene, naphthyl and fluorene groups alkyl groups such as t-butyl, heterocyclic groups such as carbazole.
- Some of the different groups  $L\alpha$  may also be the same or different charged groups such as carboxylate groups so that the group  $L_1$  can be as defined above and the groups  $L_2$ ,  $L_3$ ... can be charged groups such as



(V)

where R is R<sub>1</sub> as defined above or the groups L<sub>1</sub>, L<sub>2</sub> can be as defined above and L<sub>3</sub>... etc. are other charged groups.

R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub> can also be



where X is O, S, Se or NH.

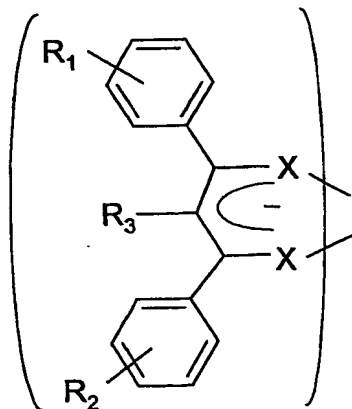
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A preferred moiety R<sub>1</sub> is trifluoromethyl CF<sub>3</sub> and examples of such diketones are, benzoyltrifluoroacetone, p-chlorobenzoyltrifluoroacetone, p-bromotrifluoroacetone, p-phenyltrifluoroacetone, 1-naphthoyltrifluoroacetone, 2-naphthoyltrifluoroacetone, 2-phenathoyltrifluoroacetone, 3-phenanthoyltrifluoroacetone, 9-anthroyltrifluoroacetone, cinnamoyltrifluoroacetone, and 2-thenoyltrifluoroacetone.

10

The different groups L<sub>α</sub> may be the same or different ligands of formulae

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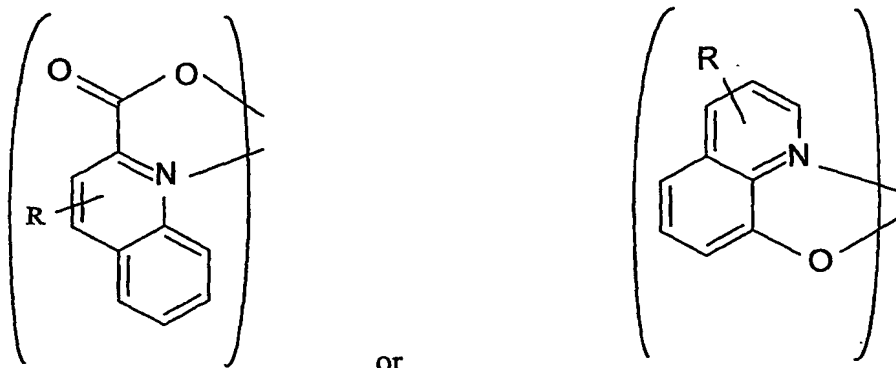


where X is O, S, or Se and R<sub>1</sub> R<sub>2</sub> and R<sub>3</sub> are as above

20

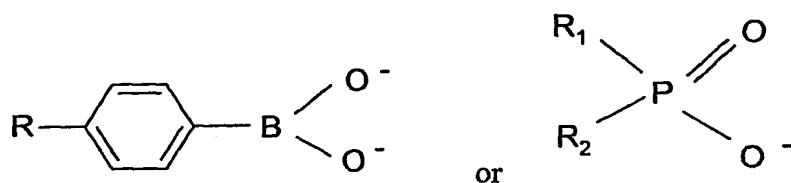
The different groups L<sub>α</sub> may be the same or different quinolate derivatives such as

- 8 -

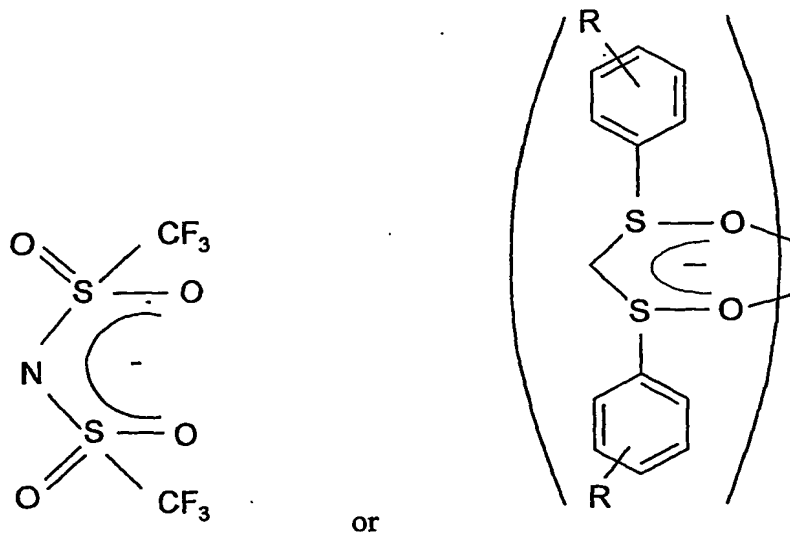


where R is hydrocarbyl, aliphatic, aromatic or heterocyclic carboxy, aryloxy, hydroxy or alkoxy e.g. the 8 hydroxy quinolate derivatives or

5

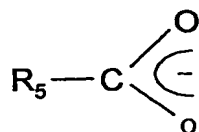


where R, R<sub>1</sub>, and R<sub>2</sub> are as above or are H or F e.g. R<sub>1</sub> and R<sub>2</sub> are alkyl or alkoxy groups .

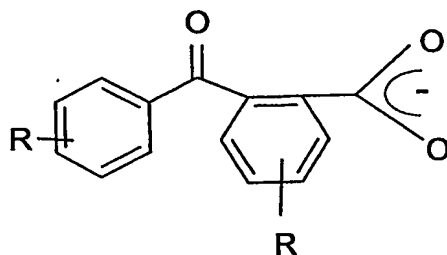


10

As stated above the different groups  $L\alpha$  may also be the same or different carboxylate groups e.g.



- 5 where  $R_5$  is a substituted or unsubstituted aromatic, polycyclic or heterocyclic ring a polypyridyl group,  $R_5$  can also be a 2-ethyl hexyl group so  $L_n$  is 2-ethylhexanoate or  $R_5$  can be a chair structure so that  $L_n$  is 2-acetyl cyclohexanoate or  $L\alpha$  can be



10

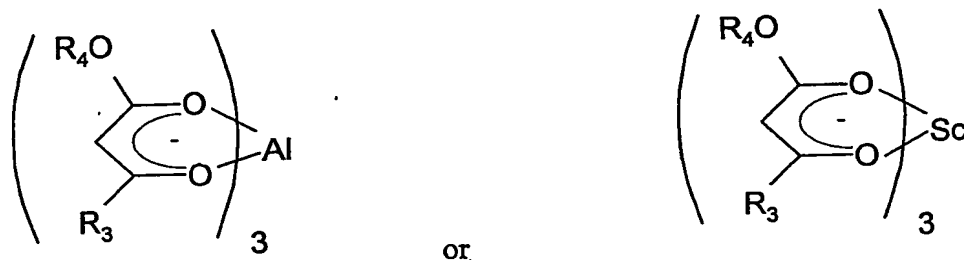
where  $R$  is as above e.g. alkyl, allenyl, amino or a fused ring such as a cyclic or polycyclic ring.

- 15 The different groups  $L\alpha$  may also be



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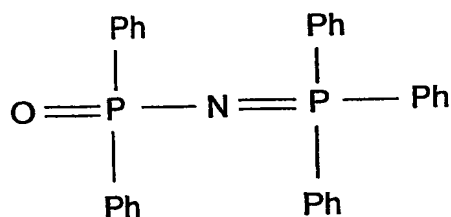
10 A preferred  $\beta$ -diketone is when  $R_1$  and/or  $R_2$  are alkoxy such as methoxy and the metals are aluminium or scandium i.e. the complexes have the formula



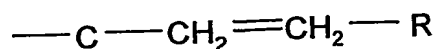
where  $R_4$  is an alkyl group, preferably methyl and  $R_3$  is hydrogen, an alkyl group such as methyl or  $R_4O$ .

The groups  $L_P$  can be selected from

5



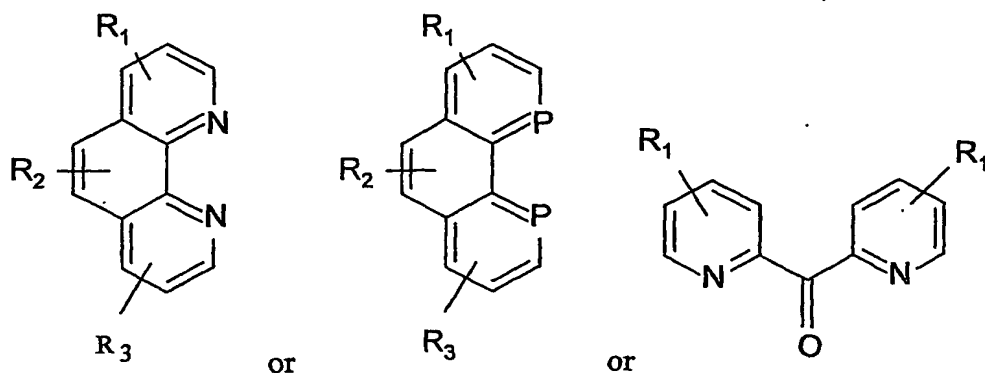
Where each Ph which can be the same or different and can be a phenyl (OPNP) or a substituted phenyl group, other substituted or unsubstituted aromatic group, a substituted or unsubstituted heterocyclic or polycyclic group, a substituted or unsubstituted fused aromatic group such as a naphthyl, anthracene, phenanthrene or pyrene group. The substituents can be for example an alkyl, aralkyl, alkoxy, aromatic, heterocyclic, polycyclic group, halogen such as fluorine, cyano, amino. Substituted amino etc. Examples are given in figs. 1 and 2 of the drawings where  $R$ ,  $R_1$ ,  $R_2$ ,  $R_3$  and  $R_4$  can be the same or different and are selected from hydrogen, hydrocarbyl groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoromethyl groups, halogens such as fluorine or thiophenyl groups;  $R$ ,  $R_1$ ,  $R_2$ ,  $R_3$  and  $R_4$  can also form substituted and unsubstituted fused aromatic, heterocyclic and polycyclic ring structures and can be copolymerisable with a monomer e.g. styrene.  $R$ ,  $R_1$ ,  $R_2$ ,  $R_3$  and  $R_4$  can also be unsaturated alkylene groups such as vinyl groups or groups



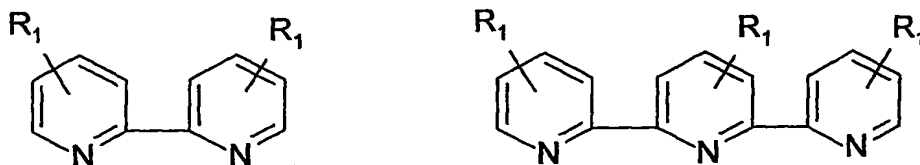
where  $R$  is as above.

25  $L_P$  can also be compounds of formulae

- 12 -



where  $R_1$ ,  $R_2$  and  $R_3$  are as referred to above, for example bathophen shown in fig. 3 of the drawings in which R is as above or

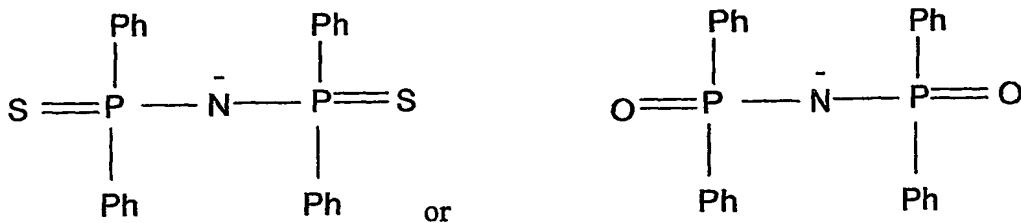


5

where  $R_1$ ,  $R_2$  and  $R_3$  are as referred to above.

$L_p$  can also be

10



where Ph is as above.

15 Other examples of  $L_p$  chelates are as shown in figs. 4 and fluorene and fluorene derivatives e.g. a shown in figs. 5 and compounds of formulae as shown as shown in figs. 6 to 8.

Specific examples of  $L\alpha$  and  $Lp$  are tripyridyl and TMHD, and TMHD complexes,  $\alpha$ ,  $\alpha'$ ,  $\alpha''$  tripyridyl, crown ethers, cyclans, cryptans phthalocyanans, porphoryins ethylene diamine tetramine (EDTA), DCTA, DTPA and TTHA. Where TMHD is  
5 2,2,6,6-tetramethyl-3,5-heptanedionato and OPNP is diphenylphosphonimide triphenyl phosphorane. The formulae of the polyamines are shown in fig. 9.

Optionally there is a layer of an electron transmitting material between the cathode and the electroluminescent material layer, the electron transmitting material is a  
10 material which will transport electrons when an electric current is passed through electron transmitting materials include a metal complex such as a metal quinolate e.g. an aluminium quinolate, lithium quinolate a cyano anthracene such as 9,10 dicyano anthracene, cyano substituted aromatic compounds such as polycyanoanthracenes, tetracyanoquinodimethane, a polystyrene sulphonate and compounds of formulae  
15 shown in fig. 10. Instead of being a separate layer the electron transmitting material can be mixed with the electroluminescent material and co-deposited with it.

Preferably there is a hole transporting layer deposited on the transparent substrate and the electroluminescent material is deposited on the hole transporting layer. The hole  
20 transporting layer serves to transport holes and to block the electrons, thus preventing electrons from moving into the electrode without recombining with holes. The recombination of carriers therefore mainly takes place in the emitter layer.

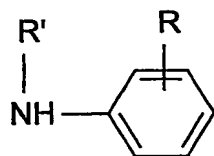
Hole transmitting layers are used in polymer electroluminescent devices and any of  
25 the known hole transmitting materials in film form can be used.

The hole transmitting layer can be made of a film of an aromatic amine complex such as poly (vinylcarbazole), N, N'-diphenyl-N, N'-bis (3-methylphenyl) -1,1' -biphenyl -  
4,4'-diamine (TPD), an unsubstituted or substituted polymer of an amino substituted  
30 aromatic compound, a polyaniline, substituted polyanilines, polythiophenes,



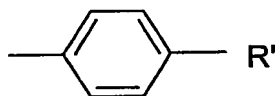
- 14 -

substituted polythiophenes, polysilanes etc. Examples of polyanilines are polymers of



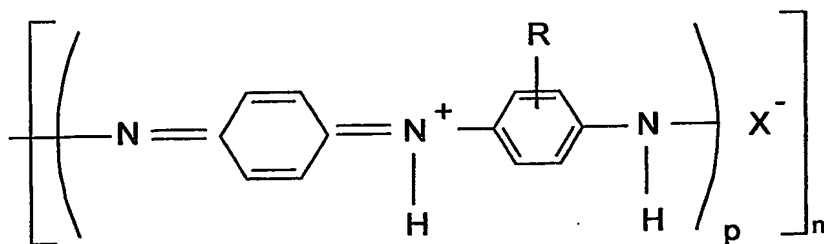
(II)

- 5 where R is in the ortho - or meta-position and is hydrogen, C1-18 alkyl, C1-6 alkoxy, amino, chloro, bromo, hydroxy or the group



- 10 where R is alkyl or aryl and R' is hydrogen, C1-6 alkyl or aryl with at least one other monomer of formula I above.

Polyanilines which can be used in the present invention have the general formula



(III)

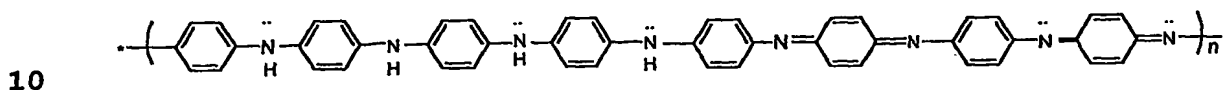
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where p is from 1 to 10 and n is from 1 to 20, R is as defined above and X is an anion, preferably selected from Cl, Br, SO<sub>4</sub>, BF<sub>4</sub>, PF<sub>6</sub>, H<sub>2</sub>PO<sub>3</sub>, H<sub>2</sub>PO<sub>4</sub>, arylsulphonate, arenedicarboxylate, polystyrenesulphonate, polyacrylate alkylsulphonate, vinylsulphonate, vinylbenzene sulphonate, cellulosesulphonate, camphor sulphonates, cellulose sulphate or a perfluorinated polyanion.

20

Examples of arylsulphonates are p-toluenesulphonate, benzenesulphonate, 9,10-anthraquinone-sulphonate and anthracenesulphonate, an example of an arenedicarboxylate is phthalate and an example of arenecarboxylate is benzoate.

- 5 A polyaniline can be formed of octamer units i.e. p is four. The conductivity of the polyaniline is dependant on the degree of protonation with the maximum conductivity being when the degree of protonation is between 40 and 60% e.g. about 50% for example



The preferred polyanilines have conductivities of the order of  $1 \times 10^{-1}$  Siemen  $\text{cm}^{-1}$  or higher.

- 15 The degree of protonation can be controlled by forming a protonated polyaniline and deprotonating. Methods of preparing polyanilines are described in the article by A. G. MacDiarmid and A. F. Epstein, Faraday Discussions, Chem Soc.88 P319 1989.

- 20 The aromatic rings can be unsubstituted or substituted e.g. by a C1 to 20 alkyl group such as ethyl.

- 25 The polyaniline can be a copolymer of aniline and preferred copolymers are the copolymers of aniline with o-anisidine, m-sulphanilic acid or o-aminophenol, or o-toluidine with o-aminophenol, o-ethylaniline, o-phenylene diamine or with amino anthracenes.

Other polymers of an amino substituted aromatic compound which can be used include substituted or unsubstituted polyaminonaphthalenes, polyaminoanthracenes, polyaminophenanthrenes, etc. and polymers of any other condensed polyaromatic

compound. Polyaminoanthracenes and methods of making them are disclosed in US Patent 6,153,726. The aromatic rings can be unsubstituted or substituted e.g. by a group R as defined above.

- 5 The polyanilines can be deposited on the first electrode by conventional methods e.g. by vacuum evaporation, spin coating, chemical deposition, direct electrodeposition etc. preferably the thickness of the polyaniline layer is such that the layer is conductive and transparent and can is preferably from 20nm to 200nm. The polyanilines can be doped or undoped, when they are doped they can be dissolved in a  
10 solvent and deposited as a film, when they are undoped they are solids and can be deposited by vacuum evaporation i.e. by sublimation.

The polymers of an amino substituted aromatic compound such as polyanilines referred to above can also be used as buffer layers with other hole transporting  
15 materials.

The structural formulae of some other hole transmitting materials are shown in Figures 11, 12, 13 and 14 of the drawings, where  $R_1$ ,  $R_2$  and  $R_3$  can be the same or different and are selected from hydrogen, and substituted and unsubstituted  
20 hydrocarbyl groups such as substituted and unsubstituted aliphatic groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoromethyl groups, halogens such as fluorine or thiophenyl groups;  $R_1$ ,  $R_2$  and  $R_3$  can also form substituted and unsubstituted fused aromatic, heterocyclic and polycyclic ring structures and can be copolymerisable with a monomer e.g.  
25 styrene. X is Se, S or O, Y can be hydrogen, substituted or unsubstituted hydrocarbyl groups, such as substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorine, fluorocarbons such as trifluoromethyl groups, halogens such as fluorine or thiophenyl groups or nitrile.

Examples of R<sub>1</sub> and/or R<sub>2</sub> and/or R<sub>3</sub> include aliphatic, aromatic and heterocyclic alkoxy, aryloxy and carboxy groups, substituted and substituted phenyl, fluorophenyl, biphenyl, phenanthrene, anthracene, naphthyl and fluorene groups alkyl groups such as t-butyl, heterocyclic groups such as carbazole.

5

Other hole transporting materials which can be used are conjugated polymers.

10 US Patent 5807627 discloses an electroluminescence device in which there are conjugated polymers in the electroluminescent layer. The conjugated polymers referred to are defined as polymers for which the main chain is either fully conjugated possessing extended pi molecular orbitals along the length of the chain or else is substantially conjugated, but with interruptions to conjugation, either random or regular along the main chain. They can be homopolymers or copolymers.

15 The conjugated polymer used can be any of the conjugated polymers disclosed or referred to in US 5807627, PCT/WO90/13148 and PCT/WO92/03490.

20 The conjugated polymers disclosed are poly (p-phenylenevinylene)-PPV and copolymers including PPV. Other preferred polymers are poly(2,5 dialkoxyphenylene vinylene) such as poly (2-methoxy-5-(2-methoxypentyloxy-1,4-phenylene vinylene), poly(2-methoxypentyloxy)-1,4-phenylenevinylene), poly(2-methoxy-5-(2-dodecyloxy-1,4-phenylenevinylene) and other poly(2,5 dialkoxyphenylenevinylenes) with at least one of the alkoxy groups being a long chain solubilising alkoxy group, poly fluorenes and oligofluorenes, polyphenylenes and oligophenylenes, 25 polyanthracenes and oligo anthracenes, ploythiophenes and oligothiophenes.

In PPV the phenylene ring may optionally carry one or more substituents e.g. each independently selected from alkyl, preferably methyl, alkoxy, preferably methoxy or ethoxy.

Any poly(arylenevinylene) including substituted derivatives thereof can be used and the phenylene ring in poly(p-phenylenevinylene) may be replaced by a fused ring system such as anthracene or naphthlyene ring and the number of vinylene groups in each polyphenylenevinylene moeity can be increased e.g. up to 7 or higher.

5

The conjugated polymers can be made by the methods disclosed in US 5807627, PCT/WO90/13148 and PCT/WO92/03490.

10

The hole transporting material can optionally be mixed with the electroluminescent material in a ratio of 5 - 95% of the electroluminescent material to 95 to 5% of the hole transporting compound.

15

The hole transporting materials, the electroluminescent material and the electron injecting materials can be mixed together to form one layer, which simplifies the construction.

20

The first electrode is preferably a transparent substrate which is a conductive glass or plastic material which acts as the cathode, preferred substrates are conductive glasses such as indium tin oxide coated glass, but any glass which is conductive or has a conductive layer can be used. Conductive polymers and conductive polymer coated glass or plastics materials can also be used as the substrate. The first electrode can comprise a transparent metal such as gold, silver a platinum group metal etc.

25

In general the thickness of the layers is from 5nm to 500nm.

30

The second electrode functions as the cathode and can be any low work function metal e.g. aluminium, calcium, lithium, silver/magnesium alloys etc., aluminium is a preferred metal.

The display of the invention may be monochromatic or polychromatic.

Electroluminescent rare earth chelate compounds are known which will emit a range of colours e.g. red, green, and blue light and white light and examples are disclosed in Patent Applications WO98/58037 PCT/GB98/01773, PCT/GB99/03619, PCT/GB99/04030, PCT/GB99/04024, PCT/GB99/04028, PCT/GB00/00268 and can  
5 be used to form OLEDs emitting those colours. Thus, a full colour display can be formed by arranging three individual backplanes, each emitting a different primary monochrome colour, on different sides of an optical system, from another side of which a combined colour image can be viewed. Alternatively, rare earth chelate electroluminescent compounds emitting different colours can be fabricated so that  
10 adjacent diode pixels in groups of three neighbouring pixels produce red, green and blue light. In a further alternative, field sequential colour filters can be fitted to a white light emitting display.

Either or both electrodes can be formed of silicon and the electroluminescent material  
15 and intervening layers of a hole transporting and electron transporting materials can be formed as pixels on the silicon substrate. Preferably each pixel comprises at least one layer of a rare earth chelate electroluminescent material and an (at least semi-) transparent electrode in contact with the organic layer on a side thereof remote from the substrate.

20 Preferably, the substrate is of crystalline silicon and the surface of the substrate may be polished or smoothed to produce a flat surface prior to the deposition of electrode, or electroluminescent compound. Alternatively a non-planarised silicon substrate can be coated with a layer of conducting polymer to provide a smooth, flat surface prior to  
25 deposition of further materials.

In one embodiment, each pixel comprises a metal electrode in contact with the substrate. Depending on the relative work functions of the metal and transparent electrodes, either may serve as the anode with the other constituting the cathode.

When the silicon substrate is the cathode an indium tin oxide coated glass can act as the anode and light is emitted through the anode. When the silicon substrate acts as the anode the cathode can be formed of a transparent electrode which has a suitable work function, for example by a indium zinc oxide coated glass in which the indium zinc oxide has a low work function. The anode can have a transparent coating of a metal formed on it to give a suitable work function. These devices are sometimes referred to as top emitting devices or back emitting devices.

The metal electrode may consist of a plurality of metal layers, for example a higher work function metal such as aluminium deposited on the substrate and a lower work function metal such as calcium deposited on the higher work function metal. In another example, a further layer of conducting polymer lies on top of a stable metal such as aluminium.

Preferably, the electrode also acts as a mirror behind each pixel and is either deposited on, or sunk into, the planarised surface of the substrate. However, there may alternatively be a light absorbing black layer adjacent to the substrate.

In still another embodiment, selective regions of a bottom conducting polymer layer are made non-conducting by exposure to a suitable aqueous solution allowing formation of arrays of conducting pixel pads which serve as the bottom contacts of the pixel electrodes.

As described in WO00/60669 the brightness of light emitted from each pixel is preferably controllable in an analogue manner by adjusting the voltage or current applied by the matrix circuitry or by inputting a digital signal which is converted to an analogue signal in each pixel circuit. The substrate preferably also provides data drivers, data converters and scan drivers for processing information to address the array of pixels so as to create images. When an electroluminescent material is used which emits light of a different colour depending on the applied voltage the colour of

each pixel can be controlled by the matrix circuitry.

5 In one embodiment, each pixel is controlled by a switch comprising a voltage controlled element and a variable resistance element, both of which are conveniently formed by metal-oxide-semiconductor field effect transistors (MOSFETs) or by an active matrix transistor.

10 US patent 4769292 disclose a range of fluorescent dyes which can be used in luminescent devices in which the electroluminescent material is aluminium quinolate or an aluminium quinolate derivative.

15 We have now discovered that fluorescent dyes can be added to electroluminescent complexes of general formula  $(L\alpha)_nM$  where M is a rare earth, lanthanide or an actinide,  $L\alpha$  is an organic complex and n is the valence state of M.

20 As explained in US 4769292 the light is emitted in the electroluminescent material layer in response to the injection and combination of holes in the layer. If a fluorescent material is added to the electroluminescent the colour of the emitted light can be varied. In theory, if an electroluminescent host material and a fluorescent material could be found for blending which have exactly the same affinity for hole-electron recombination each material should emit light upon injection of holes and electrons in the luminescent zone. The perceived hue of light emission would be the visual integration of both emissions. Since imposing such a balance of host and fluorescent materials is highly limiting, it is preferred to choose the fluorescent material so that it provides the favored sites for light emission. When only a small proportion of fluorescent material providing favored sites for light emission is present, peak intensity wavelength emissions typical of the host material can be entirely eliminated in favor of a new peak intensity wavelength emission attributable to the fluorescent material. While the minimum proportion of fluorescent material  
30 sufficient to achieve this effect varies by the specific choice of host and fluorescent



materials, in no instance is it necessary to employ more than about 10 mole percent fluorescent material, based on moles of host material and seldom is it necessary to employ more than 1 mole percent of the fluorescent material. On the other hand, for any host material capable of emitting light in the absence of fluorescent material, limiting the fluorescent material present to extremely small amounts, typically less than about  $10^{-3}$  mole percent, based on host material, can result in retaining emission at wavelengths characteristic of the host material. Thus, by choosing the proportion of a fluorescent material capable of providing favored sites for light emission, either a full or partial shifting of emission wavelengths can be realized. This allows the spectral emissions of the EL devices of this invention to be selected and balanced to suit the application to be served.

Choosing fluorescent materials capable of providing favored sites for light emission necessarily involves relating the properties of the fluorescent material to those of the host material. The host material can be viewed as a collector for injected holes and electrons with the fluorescent material providing the molecular sites for light emission. One important relationship for choosing a fluorescent material capable of modifying the hue of light emission when present in a host material is a comparison of the reduction potentials of the two materials. The fluorescent materials demonstrated to shift the wavelength of light emission have exhibited a less negative reduction potential than that of the host material.

Reduction potentials, measured in electron volts, have been widely reported in the literature along with varied techniques for their measurement. Since it is a comparison of reduction potentials rather than their absolute values which is desired, it is apparent that any accepted technique for reduction potential measurement can be employed, provided both the fluorescent and host material reduction potentials are similarly measured. A preferred oxidation and reduction potential measurement techniques is reported by R. J. Cox, Photographic Sensitivity, Academic Press, 1973, Chapter 15.

A second important relationship for choosing a fluorescent material capable of modifying the hue of light emission when present in a host material is a comparison of the bandgap potentials of the two materials. The fluorescent materials demonstrated to shift the wavelength of light emission have exhibited a lower bandgap potential than that of the host material. The bandgap potential of a molecule is taken as the potential difference in electron volts (eV) separating its ground state and first singlet state. Bandgap potentials and techniques for their measurement have been widely reported in the literature. The bandgap potentials herein reported are those measured in electron volts (eV) at an absorption wavelength which is bathochromic to the absorption peak and of a magnitude one tenth that of the magnitude of the absorption peak. Since it is a comparison of bandgap potentials rather than their absolute values which is desired, it is apparent that any accepted technique for bandgap measurement can be employed, provided both the fluorescent and host material band gaps are similarly measured.

One illustrative measurement technique is disclosed by F. Gutman and L. E. Lyons, Organic Semiconductors, Wiley, 1967, Chapter 5. Where a host material is chosen which is itself capable of emitting light in the absence of the fluorescent material, it has been observed that suppression of light emission at the wavelengths of emission characteristics of the host material alone and enhancement of emission at wavelengths characteristic of the fluorescent material occurs when spectral coupling of the host and fluorescent materials is achieved. By spectral coupling it is meant that an overlap exists between the wavelengths of emission characteristic of the host material alone and the wavelengths of light absorption of the fluorescent material in the absence of the host material. Optimal spectral coupling occurs when the maximum emission of the host material alone substantially matches within  $\pm 25$  nm the maximum absorption of the fluorescent material alone.

In practice advantageous spectral coupling can occur with peak emission and absorption wavelengths differing by up to 100 nm or more, depending on the width of the peaks and their hypsochromic and bathochromic slopes. Where less than optimum spectral coupling between the host and fluorescent materials is contemplated, a bathochromic as compared to a hypsochromic displacement of the fluorescent material produces more efficient results.

Useful fluorescent materials are those capable of being blended with the host material and fabricated into thin films satisfying the thickness ranges described above forming the luminescent zones of the EL devices of this invention. While crystalline host materials do not lend themselves to thin film formation, the limited amounts of fluorescent materials present in the host materials permits the use of fluorescent materials which are alone incapable of thin film formation.

Preferred fluorescent materials are those which form a common phase with the host material.

Fluorescent dyes constitute a preferred class of fluorescent materials, since dyes lend themselves to molecular level distribution in the host material. Although any convenient technique for dispersing the fluorescent dyes in the host materials can be undertaken, preferred fluorescent dyes are those which can be vacuum vapour deposited along with the host materials.

Assuming other criteria, noted above, are satisfied, fluorescent laser dyes are recognized to be particularly useful fluorescent materials for use in the organic EL devices of this invention. One preferred class of fluorescent dyes are fluorescent coumarin dyes. Among specifically preferred fluorescent coumarin dyes as described in US patent 4769292 the contents of which are hereby incorporated by reference.

Another preferred class of fluorescent dyes are fluorescent 4-dicyanomethylene-4H-

pyrans and 4-dicyanomethylene-4H-thiopyrans, hereinafter referred to as fluorescent dicyanomethylenepyran and thiopyran dyes the preferred fluorescent dyes of this class are specified in US Patent 4769292. Useful fluorescent dyes can also be selected from among known polymethine dyes, which include the cyanines, merocyanines, complex cyanines and merocyanines (i.e., tri-, tetra and poly-nuclear cyanines and merocyanines}, oxonols, hemioxonols, styryls, merostyryls, and streptocyanines.

The cyanine dyes include, joined by a methine linkage, two basic heterocyclic nuclei, such as azolium or azinium nuclei, for example, those derived from pyridinium, quinolinium, isoquinolinium, oxazolium, thiazolium, selenazolium, indazolium, pyrazolium, pyrrolium, indolium, 3H-indolium, imidazolium, oxadiazolium, thiadioxazolium, benzoxazolium, benzothiazolium, benzoselenazolium, benzotellurazolium, benzimidazolium, 3H or 1 H-benzoindolium, naphthoxazolium, naphthothiazolium, naphthoselenazolium, naphthotellurazolium, carbazolium, pyrrolopyridinium, phenanthrothiazolium, and acenaphthothiazolium quaternary salts.

Cyanine dyes can contain two heterocyclic nuclei joined by a methine linkage containing an uneven number of methine groups or can contain a heterocyclic nucleus joined by a methine linkage containing an even number of methine groups.

The greater the number of the methine groups linking nuclei in the polymethine dyes in general and the cyanine dyes in particular the longer the absorption wavelengths of the dyes. For example, dicarbocyanine dyes (cyanine dyes containing five methine groups linking two basic heterocyclic nuclei) exhibit longer absorption wavelengths than carbocyanine dyes (cyanine dyes containing three methine groups linking two basic heterocyclic nuclei) which in turn exhibit longer absorption wavelengths than simple cyanine dyes (cyanine dyes containing a single methine group linking two basic heterocyclic nuclei). Carbocyanine and dicarbocyanine dyes are longer

wavelength dyes while simple cyanine dyes are typically yellow dyes, but can exhibit absorption maxima up to about 550 nm in wavelength with proper choice of nuclei and other components capable of bathochromically shifting absorption. Preferred polymethine dyes, particularly cyanine dyes, for use as fluorescent dyes are so-called  
5 rigidized dyes. These dyes are constructed to restrict the movement of one nucleus in relation to another. This avoids radiationless, kinetic dissipation of the excited state energy. One approach to rigidizing the dye structure is to incorporate a separate bridging group providing a separate linkage in addition to the methine chain linkage joining the terminal nuclei of the dye. Bridged polymethine dyes are illustrated by  
10 Brooker et al U.S. Pat. No. 2478,367, Brooker U.S. Pat. No. 2479,152, Gilbert U.S. Pat. No. 4,490,463, and Tredwell et al, "Picosecond Time Resolved Fluorescence Lifetimes of the Polymethine and Related Dyes", Chemical Physics, Vol. 43 (1979) pp. 307-316. The methine chain joining polymethine dye nuclei can be rigidized by including the methine chain as part of a cyclic nucleus joining the terminal basic  
15 nuclei of the dye. One of the techniques for both rigidizing and bathochromically shifting the absorption maxima of polymethine dyes in general and cyanine dyes in particular is to include in the methine linkage an oxocarbon bridging nucleus

Another useful class of fluorescent dyes are 4-oxo-4H-benz-[d,e] anthracenes,  
20 hereinafter referred to as oxobenzanthracene dyes.

Dyes of this class and their preparations are disclosed in Goswami et al U.S. Ser. No. 824,765, filed Jan. 31, 1986, commonly assigned, titled Fluorescent Dyes and Biological and Analytical uses thereof.

25 The oxobenzanthracene dyes can be prepared by known methods e.g. (1) preparation of a dihydrophenalenone by the procedure described by Cooke et al, Australian J. Chem., 11, pp. 230-235 (1958), (2) preparation of the lithium enolate of the dihydrophenalenone, (3) reaction of the lithium enolate with the appropriate  
30 phosphonium iodide reagent, and (4) reaction of this product with cupric chloride and

lithium chloride to produce the chlorinated or unchlorinated dye.

5 The oxobenzanthracene can have one or more substituents in the structure as long as the substituents do not adversely affect the fluorescence of the compound, such as alkyl (e.g., alkyl of 1 to 5 carbon atoms), aryl (e.g., phenyl), and other groups.

Another useful class of fluorescent dyes are xanthene dyes. One particularly preferred class of xanthene dyes are rhodamine dyes.

10 Another specifically preferred class of xanthene dyes are fluorescein dyes

15 Another useful group of fluorescent dyes are pyrylium, thiapyrylium, selenapyrylium, and telluropirylium dyes. Dyes from the first three of these classes are disclosed by Light U.S. Pat. No. 3,615,414 while dyes of the latter class are disclosed by Detty U.S. Pat. No. 4,584,258, the disclosures of which are here incorporated by reference. Since the latter two classes of dyes are bathochromically shifted toward the infrared the former two classes of dyes are preferred for achieving visible light emissions.

20 Another useful class of fluorescent dyes are fluorescent carbostyryl dyes. These dyes are characterized by a 2-quinolinol or isoquinolinol ring structure, often fused with other rings. The wavelength of maximum fluorescence generally increases with the presence of other fused rings.

25 Examples of more complex fused ring carbostyryl dyes are provided by Kadhim and Peters, "New Intermediates and Dyes for Synthetic Polymer Fibres Substituted Benzimidazolothioxanthenoisoquinolines for Polyester Fibres", JSDC, June 1974, pp. 199-201, and Arient et al, "Imidazole Dyes XX-Colouring Properties of 1,2-Napthooxylenebenzimidazole Derivatives", JSDC, June 1968, pp. 246-251.

Among other fused ring fluorescent dyes the perylene dyes, characterized by a dinaphthylene nucleus. A variety of useful fluorescent perylene dyes are known, such as, for example those disclosed by Rademacher et al, "Soluble Perylene Fluorescent Dyes with Photostability", Chem. Ber. , Voi. 115, pp. 2927 -2934, 1982, and  
5 European Patent Application No. 553,363A 1, published July 7, 1982.

Many other classes of known fluorescent dyes, such as acridine dyes; bis(styryl)benzene dyes; pyrene dyes; oxazine dyes; and phenyleneoxide dyes, sometimes referred to as POPOP dyes; are useful.

10

Not only are there many available classes of fluorescent dyes to choose from, there are wide choices of individual dye properties within any given class. The absorption maxima and reduction potentials of individual dyes can be varied through the choice of substituents. As the conjugation forming the chromophore of the dye is increased  
15 the absorption maximum of a dye can be shifted bathochromically.

Emission maxima are bathochromic to the absorption maxima.

Although the degree of bathochromic shifting can vary as a function of the dye class, usually the wavelength of maximum emission is from 25 to 125 nm bathochromically  
20 shifted as compared to the wavelength of maximum absorption. Thus, dyes which exhibit absorption maxima in the near ultraviolet in almost all cases exhibit maximum emissions in the blue portion of the spectrum. Dyes which exhibit absorption maxima in the blue portion of the spectrum exhibit emission maxima in the green portion of the spectrum, and, similarly, dyes with absorption maxima in the  
25 red portion of the spectra tend to exhibit emission maxima in the near infrared portion

A further class of fluorescent compounds which can be used are fluorescent dyes having a chromophoric unit containing at least 5 fused carbocyclic aromatic rings  
30 (hereinafter referred to as a pentacarbocyclic aromatic fluorescent dye). Suitable dyes

are described in US Patents 5150006 and 5405709.

These pentacarbocyclic aromatic fluorescent dyes have been discovered to be highly advantageous for reducing the wavelength of organic EL device emission. To function in a first category arrangement it is essential that the fluorescent dye absorb at a wavelength corresponding to an emission wavelength of the host compound. On the other hand, it is recognized that all fluorescent dyes emit at a longer wavelength than they absorb. Stated another way, a dye cannot emit light of a higher energy level than it absorbs. The difference between the longest wavelength absorption maxima (hereinafter referred to as the peak absorption) and the shortest wavelength emission maxima (hereinafter referred to as the peak emission) of a fluorescent dye is known as its Stokes shift. If the Stokes shift of a fluorescent dye is large, it is difficult to achieve efficient spectral coupling and still achieve peak emission at a shorter wavelength than that of the EL compound. Pentacarbocyclic aromatic fluorescent dyes are particularly suited for shifting organic EL device emissions to shorter blue wavelengths, since they exhibit Stokes shifts of from 80 nm to less than 20 nm, attributable to their relatively rigid chromophoric units. Thus, a hypsochromic shift in organic EL device emission can be realized even though the absorption peak of the pentacarbocyclic aromatic fluorescent dye is only 20 nm shorter in wavelength than the emission peak of the charge carrier compound. Preferred pentacarbocyclic aromatic fluorescent dyes are those that exhibit an absorption peak at wavelengths ranging from 100 to 20 nm shorter than the emission peak exhibited by the formula II charge carrier compound.

The pentacarbocyclic aromatic fluorescent dyes contemplated each contain at least 5 fused carbocyclic aromatic rings, which form a chromophoric unit. Fused aromatic carbocyclic rings in addition to the 5 required fused rings do not detract from performance characteristics. Preferred chromophoric units contain a perylene, benzopyrene, benzo-chrysene, benzonaphthacene, picene, pentaphene, pentacene, hexacene or anthanthrene nucleus, as the entire nucleus or fused with other aromatic



rings to complete the nucleus. Typically these dyes contain from 20 to 40 ring carbon atoms.

5 These pentacarbocyclic aromatic rings have the advantage that they can be deposited by vacuum vapour deposition, similarly as the other components of the organic medium. Since the pentacarbocyclic aromatic rings represent chromophores in and of themselves, it is not necessary that other ring substituents be present. However, many dyes containing pentacarbocyclic aromatic rings as chromophores are conventional, having been originally prepared for use in solution chemistry and therefore having substituents intended to modify solubility and, in some instances, hue.

10

When fluorescent pentacarbocyclic aromatic dyes are incorporated in a host charge acceptor compound, only a small amount of the fluorescent dye is required to realize advantages. Fluorescent pentacarbocyclic aromatic dyes are preferably incorporated in a concentration ranging from 0.05 to 5 mole percent, based on the moles of charge  
15 accepting compound. A specifically preferred concentration range is from 0.2 to 3 mole percent, based on the moles of charge accepting compound, with a concentration range of from 0.5 to 2 mole percent, based on the moles of charge accepting compound, being in most instances optimum.

20 A device according to one embodiment of the invention is shown in Fig. 15 the accompanying drawing which shows schematically a structure of the invention and in which there is a aluminium cathode (1), on which there is a layer of a electron transporting material (2), a layer of a rare earth chelate electroluminescent material incorporating a fluorescent dye (3), a layer of a hole transmitting material (4) and an  
25 anode which is a transparent ITO layer (5). When an electric field is applied between the substrate and the ITO light is emitted via (5).

1. An electroluminescent device comprising in sequence, (i) an anode, (ii) a layer of an electroluminescent material of general formula  $(L\alpha)_nM$  where M is a rare earth, lanthanide or an actinide,  $L\alpha$  is an organic complex as specified herein and n is the valence state of M, and (iii) a cathode, in which the layer of an electroluminescent material includes a fluorescent dye.

2. An electroluminescent device as claimed in claim 1 in which the electroluminescent material is of general formula

10



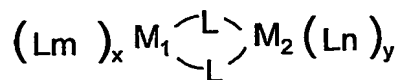
- where  $L\alpha$  and  $L_p$  are organic ligands, M is a rare earth, transition metal, lanthanide or an actinide and n is the valence state of the metal M, the ligands  $L\alpha$  can be the same or different and there can be a plurality of ligands  $L_p$  which can be the same or different where  $L\alpha$  and  $L_p$  are as specified herein.

3. An electroluminescent device as claimed in claim 1 in which the electroluminescent material of general formula  $(L\alpha)_nM_1M_2$  where  $M_1$  is the same as M above,  $M_2$  is a non rare earth metal,  $L\alpha$  is as specified herein and n is the combined valence state of  $M_1$  and  $M_2$ .

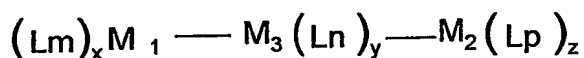
4. An electroluminescent device as claimed in claim 1 or 2 in which the complex also comprises one or more neutral ligands  $L_p$  and the complex has the general formula  $(L\alpha)_n M_1 M_2 (L_p)$ .

5. An electroluminescent device as claimed in claim 1 in which the electroluminescent material is a binuclear, trinuclear and polynuclear organometallic complexes of formula  $(Lm)_x M_1 \leftarrow M_2 (Ln)_y$  or

- 32 -

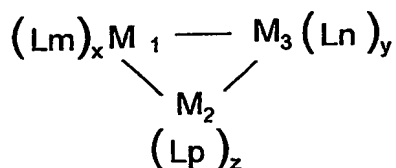


where L is a bridging ligand and where  $M_1$  is a rare earth metal and  $M_2$  is  $M_1$  or a non rare earth metal, Lm and Ln are the same or different organic ligands La as defined above, x is the valence state of  $M_1$  and y is the valence state of  $M_2$  or



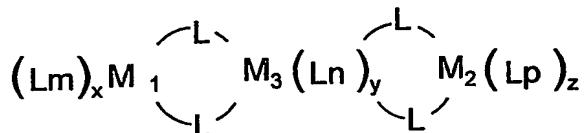
or

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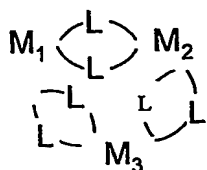


where  $M_1$ , is a rare earth metal and  $M_2$  and  $M_3$  are  $M_1$  or a non rare earth metal; Lm, Ln and Lp are organic ligands La and x is the valence state of  $M_1$ , y is the valence state of  $M_2$  and z is the valence state of  $M_3$  and Lp can be the same as Lm and Ln or different or

15



20 or



5 where L is a bridging ligand and in which the rare earth metals and the non rare earth metals can be joined together by a metal to metal bond and/or via an intermediate bridging atom, ligand or molecular group or in which there are more than three metals joined by metal to metal bonds and/or via intermediate ligands

6. An electroluminescent device as claimed in any one of claims 3 to 5 in which the metal M<sub>2</sub> is any metal which is not a rare earth, transition metal, lanthanide or an actinide.

10

7. An electroluminescent device as claimed in any one of claims 3 to 6 in which the metal M<sub>2</sub> is selected from lithium, sodium, potassium, rubidium, caesium, beryllium, magnesium, calcium, strontium, barium, copper (I), copper (II), silver, gold, zinc, cadmium, boron, aluminium, gallium, indium, germanium, tin (II), tin (IV), antimony (II), antimony (IV), lead (II), lead (IV) and metals of the first, second and third groups of transition metals in different valence states e.g. manganese, iron, ruthenium, osmium, cobalt, nickel, palladium(II), palladium(IV), platinum(II), platinum(IV), cadmium, chromium, titanium, vanadium, zirconium, tantalum, molybdenum, rhodium, iridium, titanium, niobium, scandium and yttrium.

20

8. An electroluminescent device as claimed in any preceding claim, in which there is an organic hole transporting material in contact with the layer of light emitting material.

25

9. An electroluminescent device as claimed in claim 8 in which the hole transmitting material is a film of a polymer selected from poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl) -1,1' -biphenyl -4,4'-diamine (TPD), polyaniline,

substituted polyanilines, polythiophenes, substituted polythiophenes, polysilanes and substituted polysilanes.

10. An electroluminescent device as claimed in claim 8 or 9 in which the hole  
5 transmitting material is a film of a compound of formula (II) or (III) herein or as in Figure 11, 12, 13, or 14 of the drawings.

11. An electroluminescent device as claimed in any one of claims 1 to 10 in which  
10 there is a layer of an electron transmitting material between the cathode and the electroluminescent material layer.

12. An electroluminescent device as claimed in claim 11 in which an electron  
15 transmitting material and the light emitting metal compound are mixed to form one layer.

13. An electroluminescent device as claimed in claim 11 or 12 in which the electron  
transmitting material is a metal quinolate.

14. An electroluminescent device as claimed in claim 13 in which the metal quinolate  
20 is an aluminium quinolate or lithium quinolate

15. An electroluminescent device as claimed in claim 11 or 12 in which the electron  
transmitting material is selected from cyanoanthracenes such as 9,10  
dicyanoanthracenes, polystyrene sulphonates or a compound of formulae shown in  
25 Fig. 10.

16. An electroluminescent device as claimed in any one of claims 8 to 15 in which a  
hole transmitting material and an electron transmitting material and the light emitting  
metal compound are mixed to form one layer.

17. An electroluminescent device as claimed in any one of the preceding claims in which the second electrode is selected from aluminium, calcium, lithium, silver/magnesium alloys
- 5 18. An electroluminescent device as claimed in any one the preceding claims in which the fluorescent dye has a bandgap no greater than that of the electroluminescent material and a reduction potential less negative than that of the electroluminescent material.
- 10 19. An electroluminescent device as claimed in any one the preceding claims in which the fluorescent dye is chosen from the class consisting of coumarin, dicyanomethylenepyrans and thiopyrans, polymethine, oxabenzanthracene, xanthene, pyrylium and thiapyrylium, carbostyryl, and perylene fluorescent dyes.
- 15 20. An electroluminescent device as claimed in any one of in any one the preceding claims in which the electroluminescent material is capable of emitting light of a first wavelength in the absence of said fluorescent material and said fluorescent material is capable of absorbing light at the first wavelength.
- 20 21. An electroluminescent device according to claim 20 in which the wavelength of maximum light emitted by said host material in the absence of said fluorescent material is within 25 nm of the wavelength of maximum light absorption by said fluorescent material.
- 25 22. An electroluminescent device as claimed in any one of in any one the preceding claims in which the fluorescent dye is a blue emitting dye.
23. An electroluminescent device as claimed in any one of in any one the preceding claims in which the fluorescent dye exhibits a shorter wavelength emission peak than
- 30 the electroluminescent material

24. An electroluminescent device as claimed in claim 23 in which the blue emitting fluorescent dye contains a stabilizing aromatic chromophoric unit containing at least 5 fused carbocyclic aromatic rings.

5

25. An electroluminescent device as claimed in claim 23 in which the chromophoric unit contains from 20 to 40 ring carbon atoms.

10

26. An electroluminescent device as claimed in claim 24 or 25 in which the fluorescent dye chromophoric unit is chosen from among those containing a perylene, benzopyrene, benzo-chrysene, benzonaphthacene, picene, pentaphene, pentacene, hexacene or anthanthrene nucleus.

15

27. An electroluminescent device as claimed in any one the preceding claims in which the fluorescent dye is present in a concentration ranging from 0.05 to 5 mole percent.

20

28. An electroluminescent device as claimed in any one the preceding claims in which the fluorescent dye is present in a concentration ranging from 0.2 to 3 mole percent.

- 37 -

### Abstract

An electroluminescent device comprising an electroluminescent layer which incorporates a fluorescent dye.



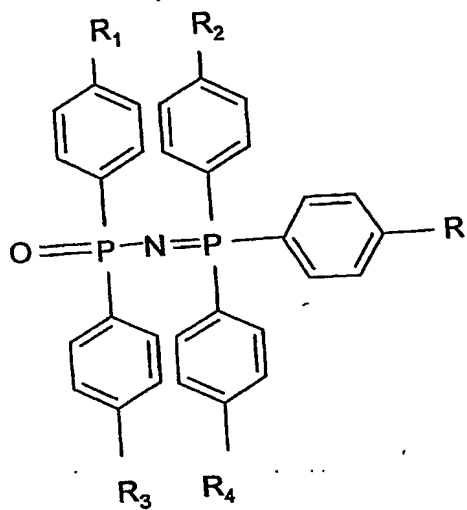


Fig. 1

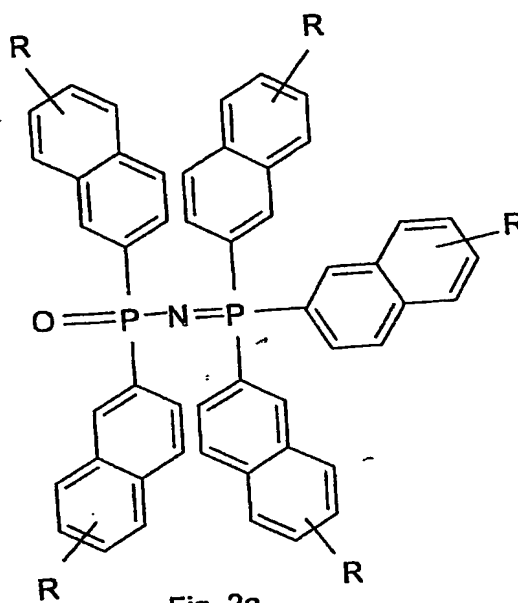


Fig. 2a

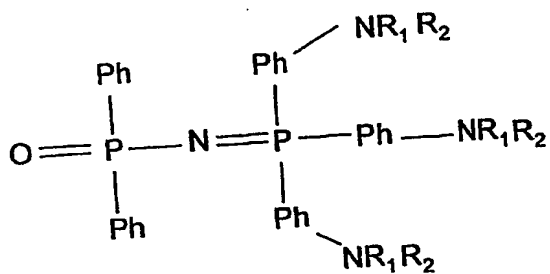


Fig. 2b

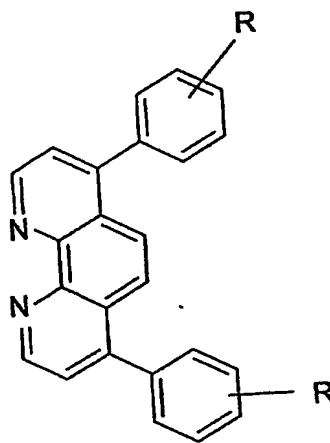


Fig. 3

2/15

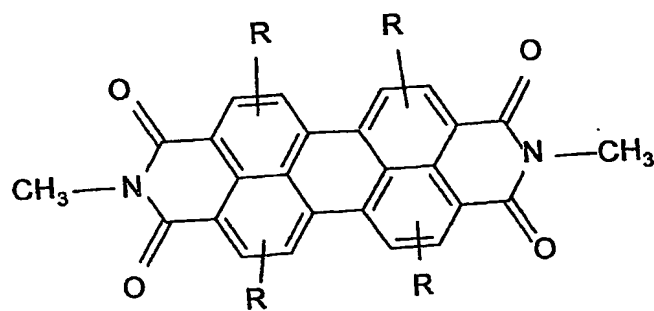


Fig. 4a

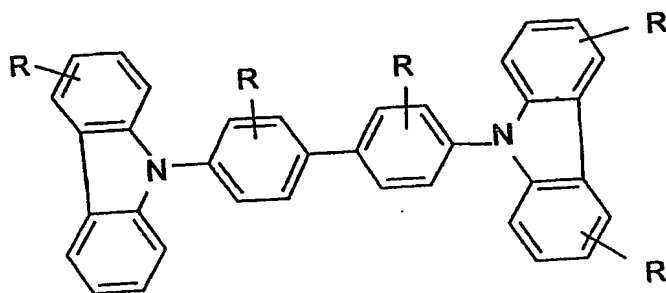


Fig. 4b

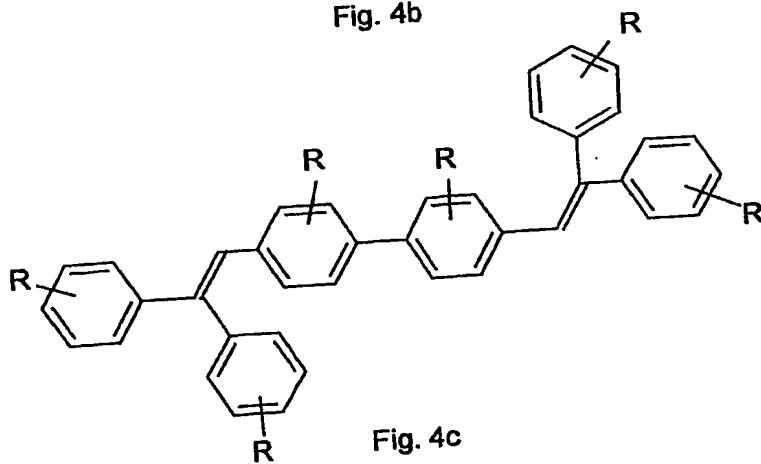


Fig. 4c

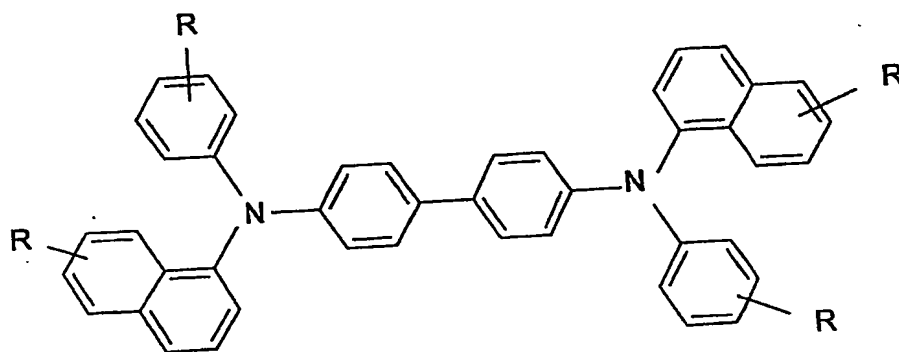


Fig. 4d

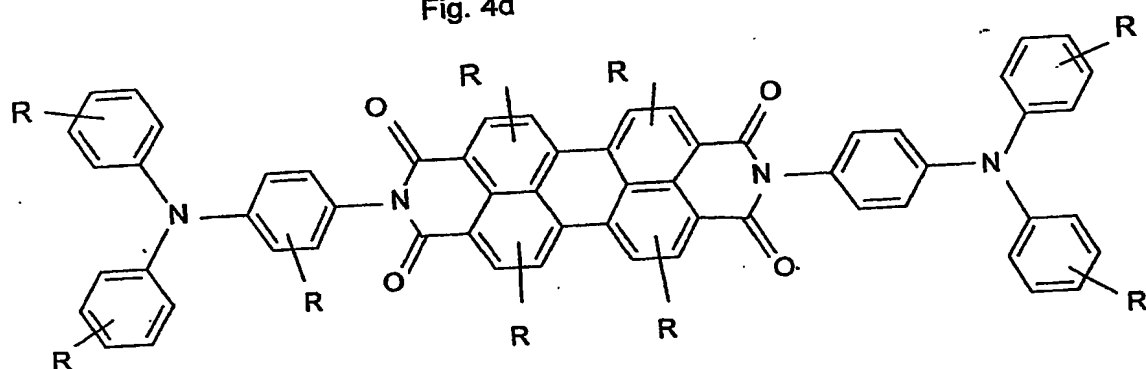


Fig. 4e

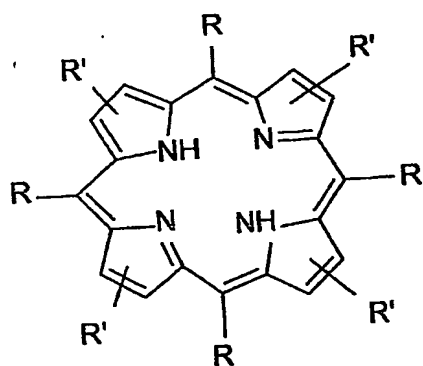


Fig. 4f

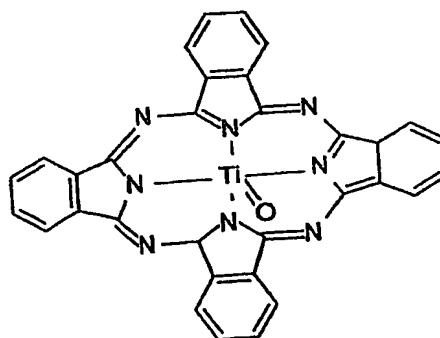


Fig. 4g

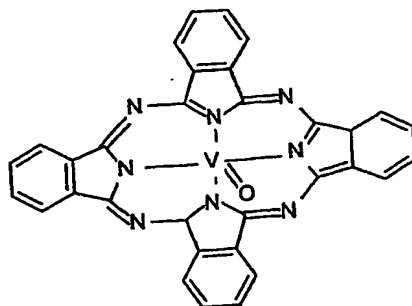


Fig. 4h

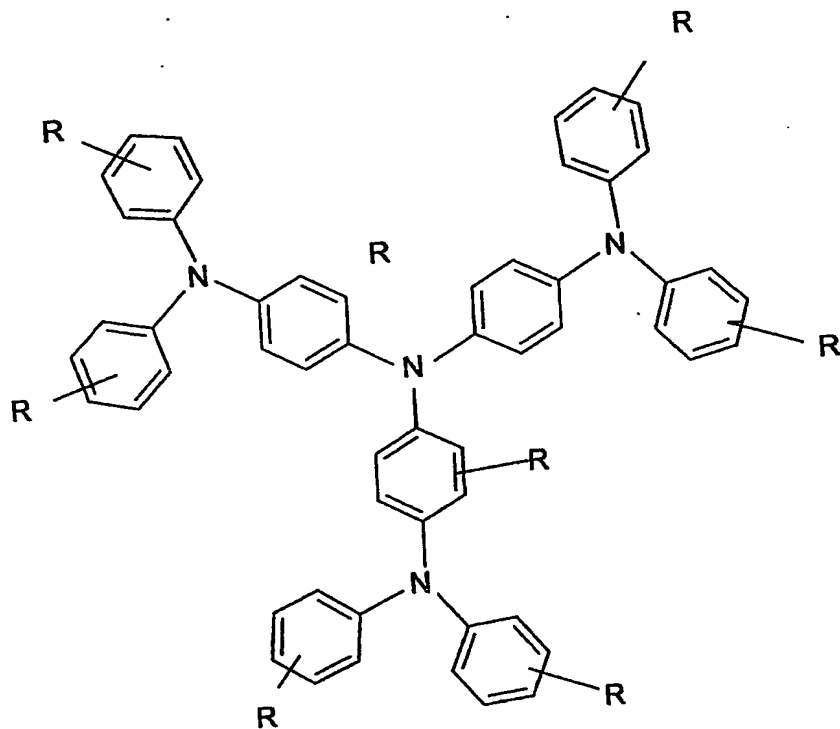


Fig. 4i

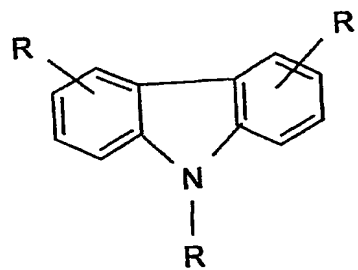


Fig. 4j

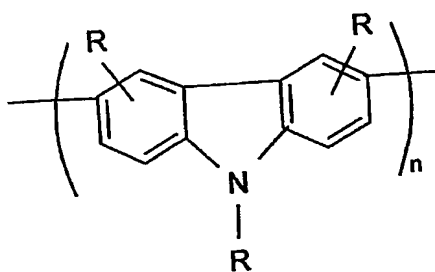


Fig. 4k

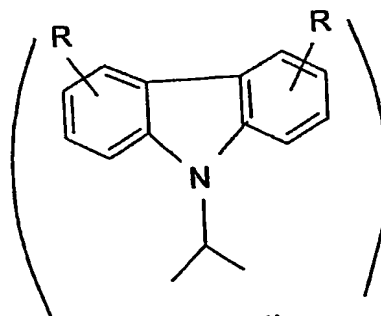


Fig. 4l

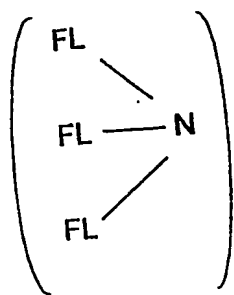


Fig. 5a

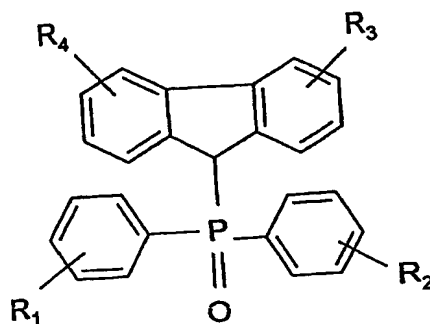


Fig. 5b

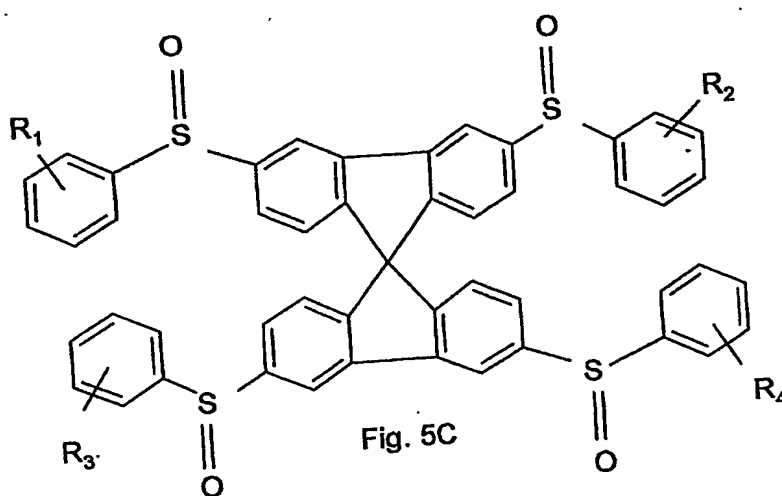


Fig. 5c

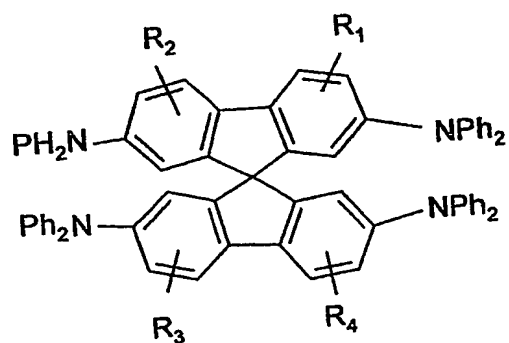


Fig. 5d

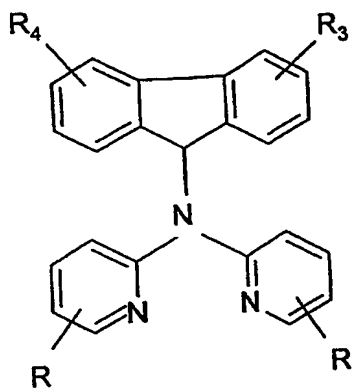


Fig. 5f

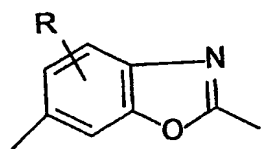


Fig. 6a

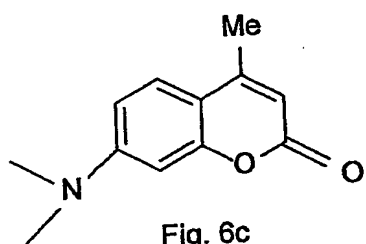


Fig. 6c

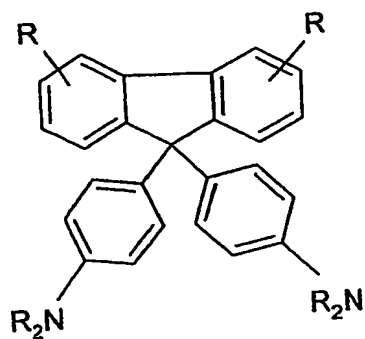


Fig. 5g

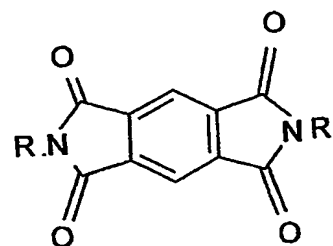


Fig 6b

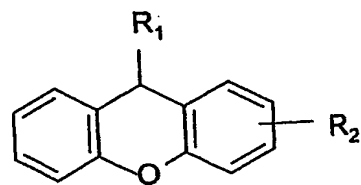


Fig. 6d

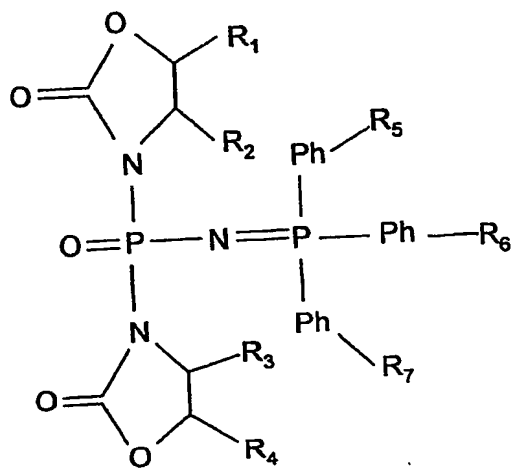


Fig. 6e

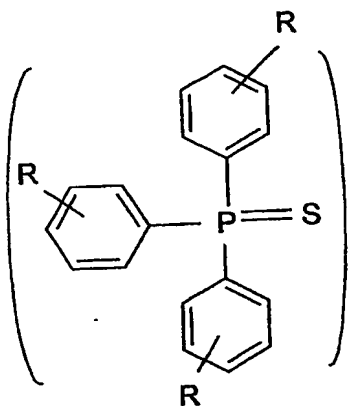


Fig. 7a

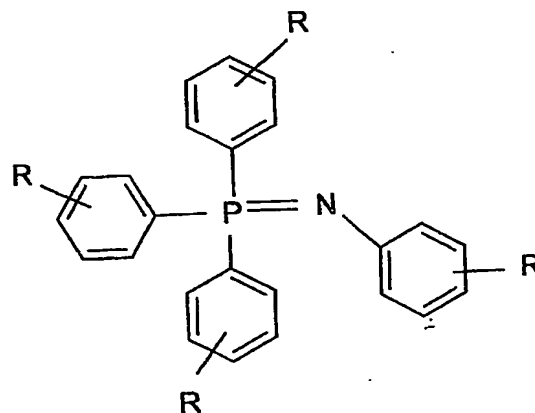


Fig. 7b

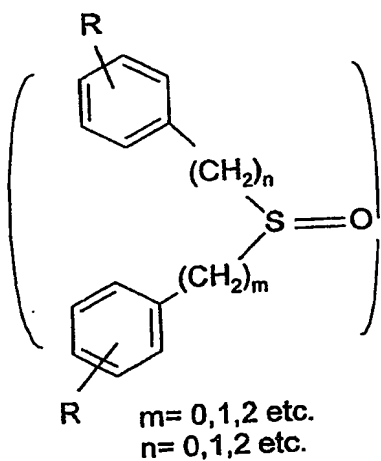


Fig. 7c

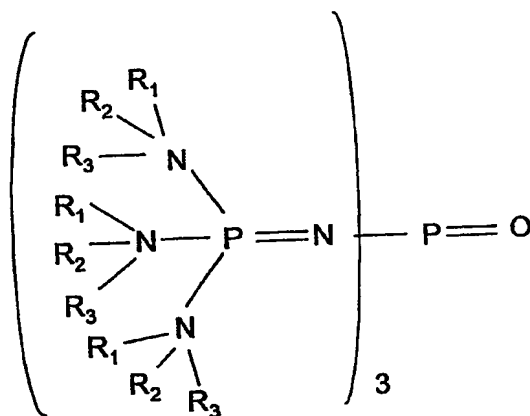


Fig. 7d

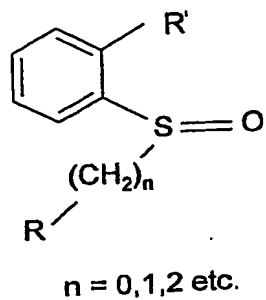


Fig. 7e

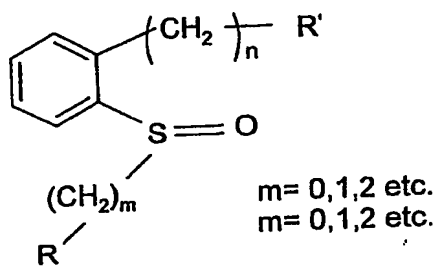


Fig. 7f

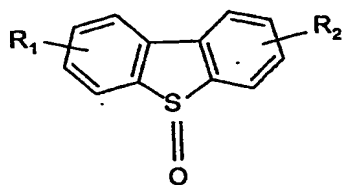


Fig. 8a

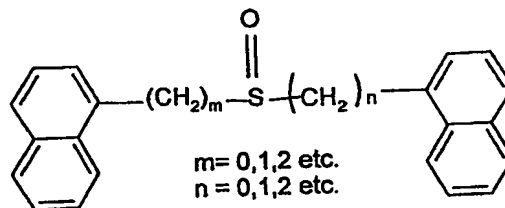


Fig. 8b

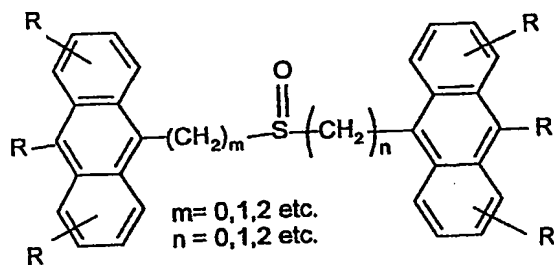


Fig. 8c

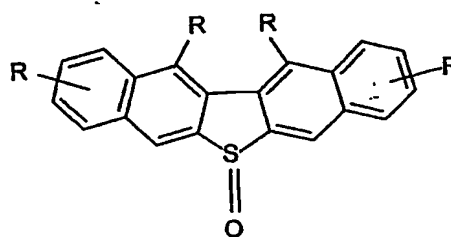


Fig. 8d

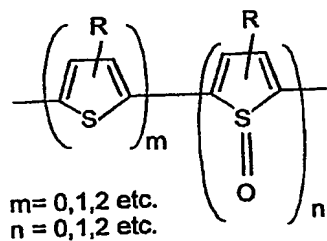


Fig. 8e

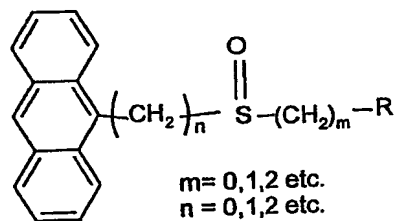


Fig. 8f

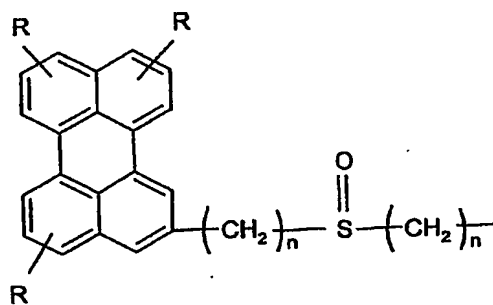


Fig. 8g

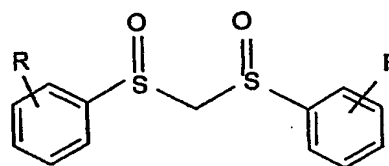
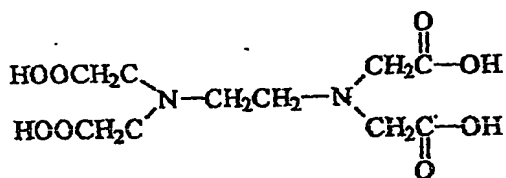


Fig. 8g



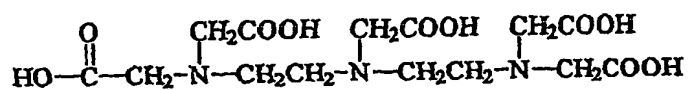
9/15



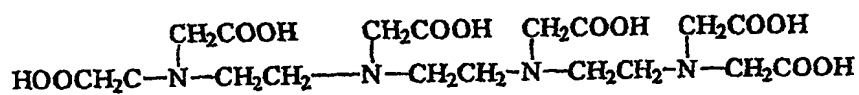
EDTA



DCTA



DTPA



TTHA

Fig, 9

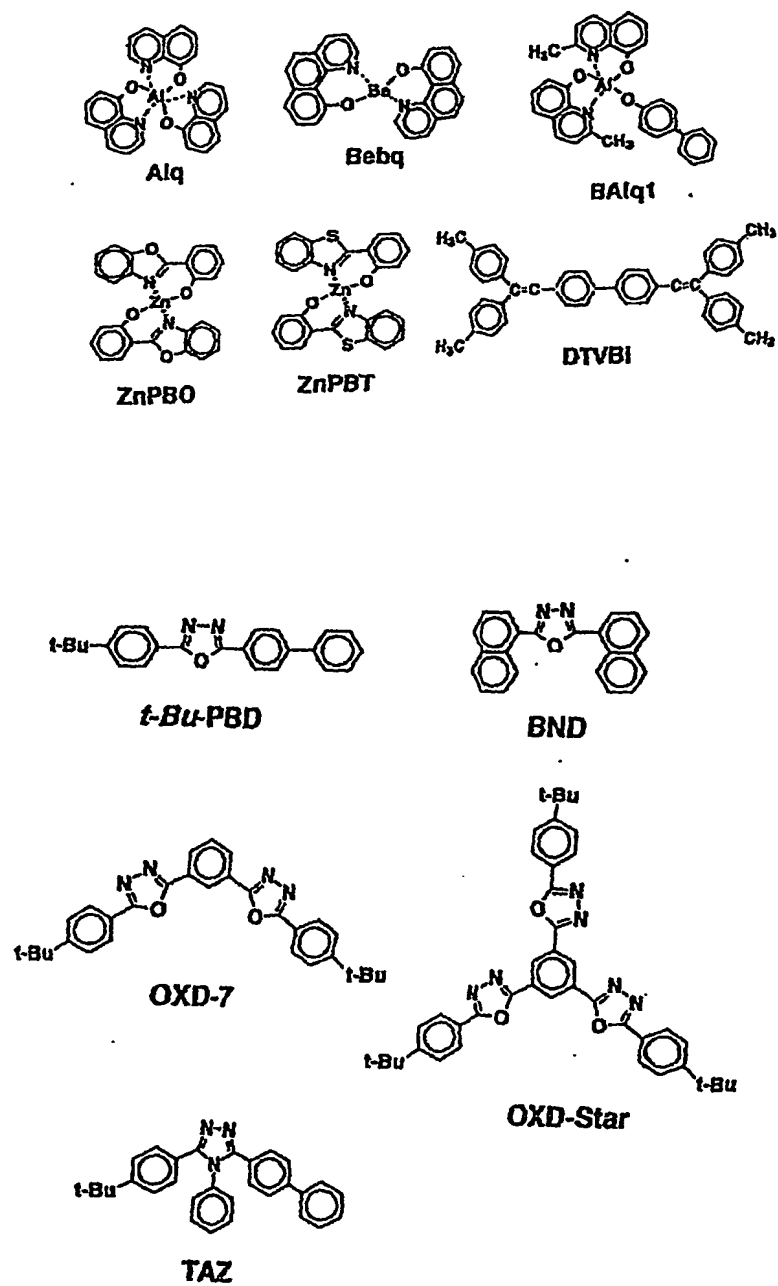


Fig. 10

11/15

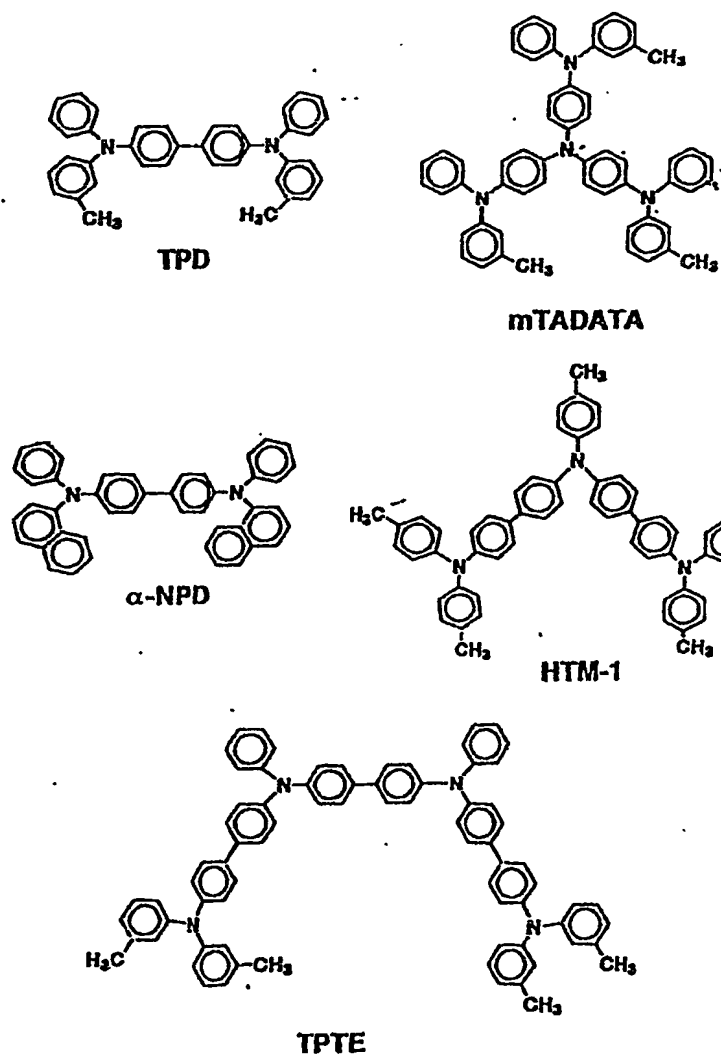


Fig. 11

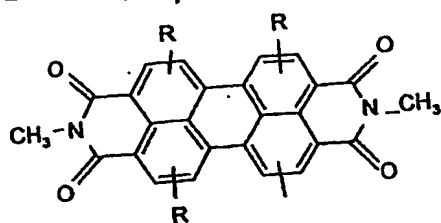


Fig. 12a

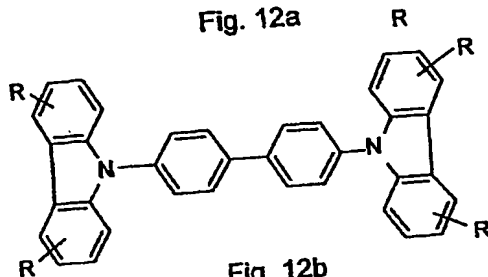


Fig. 12b

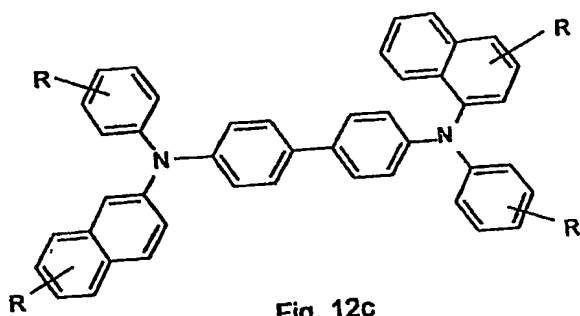


Fig. 12c

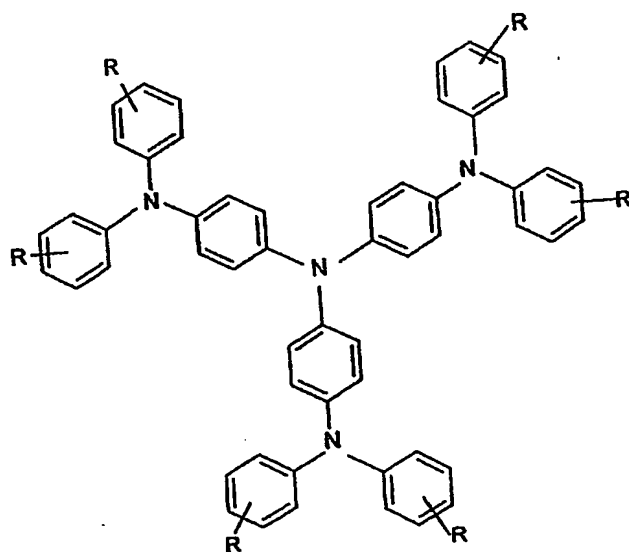


Fig. 12d

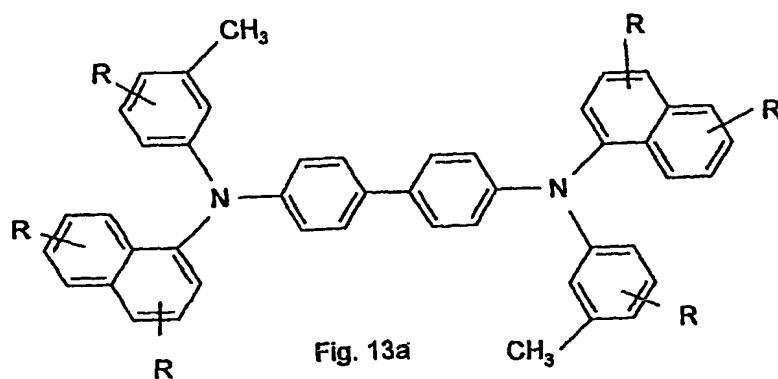


Fig. 13a

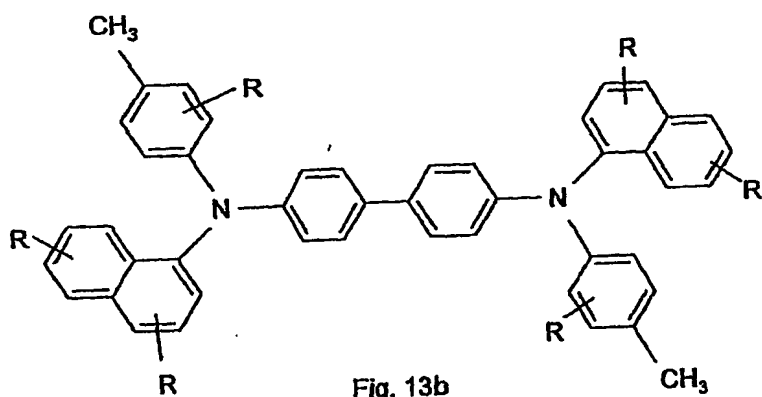


Fig. 13b

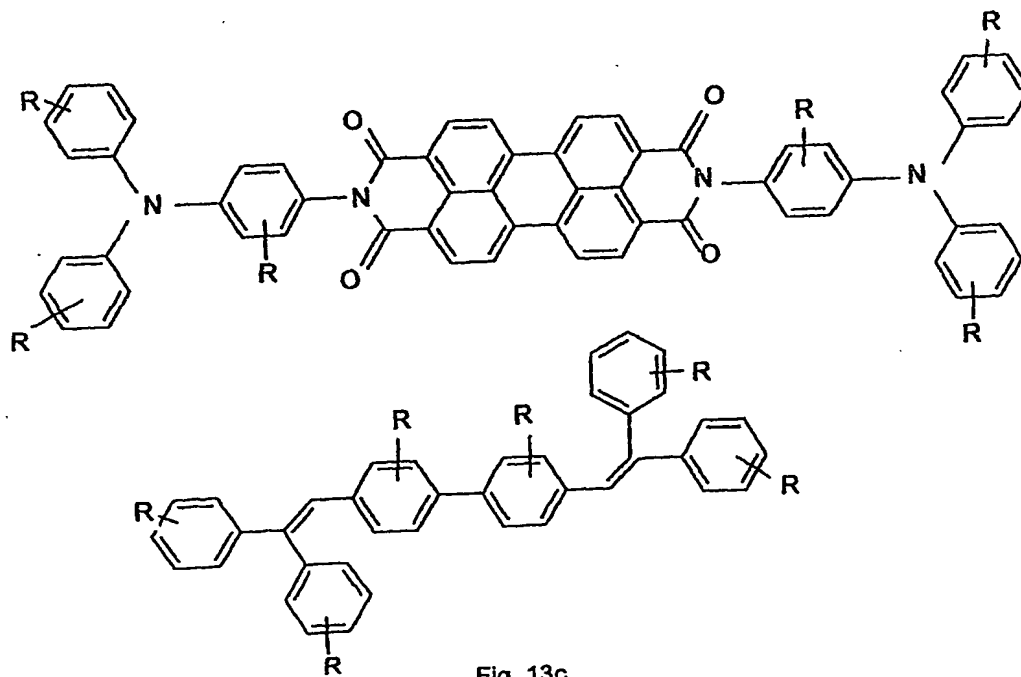


Fig. 13c

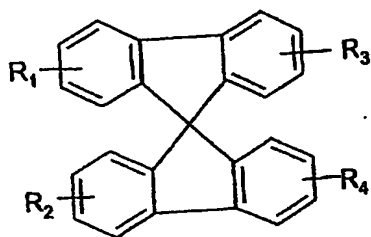


Fig. 14a

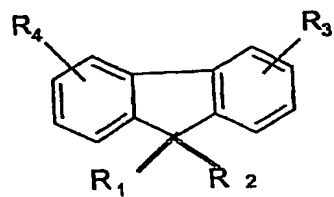
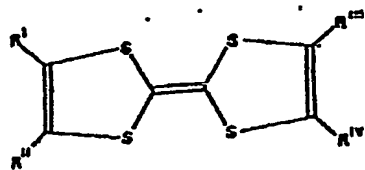


Fig. 14b



or

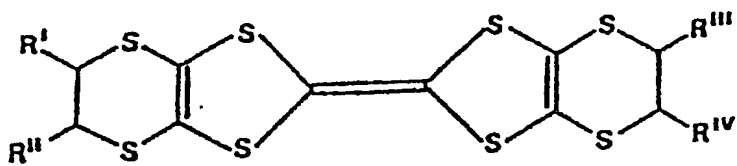


Fig. 14c

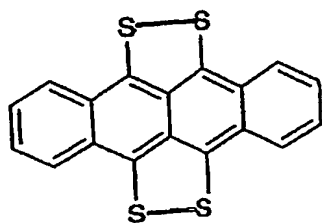


Fig. 14d

ITO / DFDA(5nm) / M-MTDATA(30nm) / Yellow(55nm) / LiF(0.7nm) / Al(150nm)  
11-1337

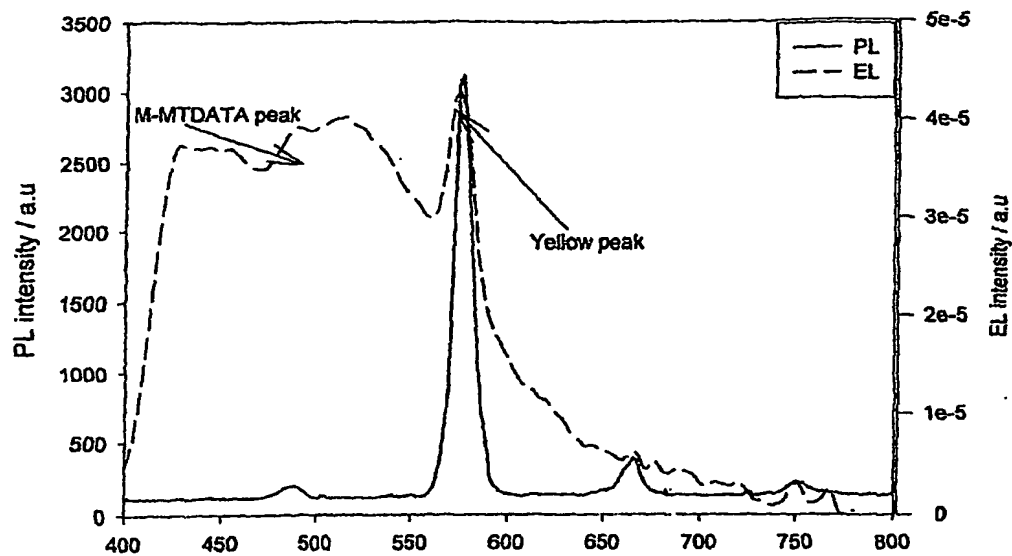


Fig. 15

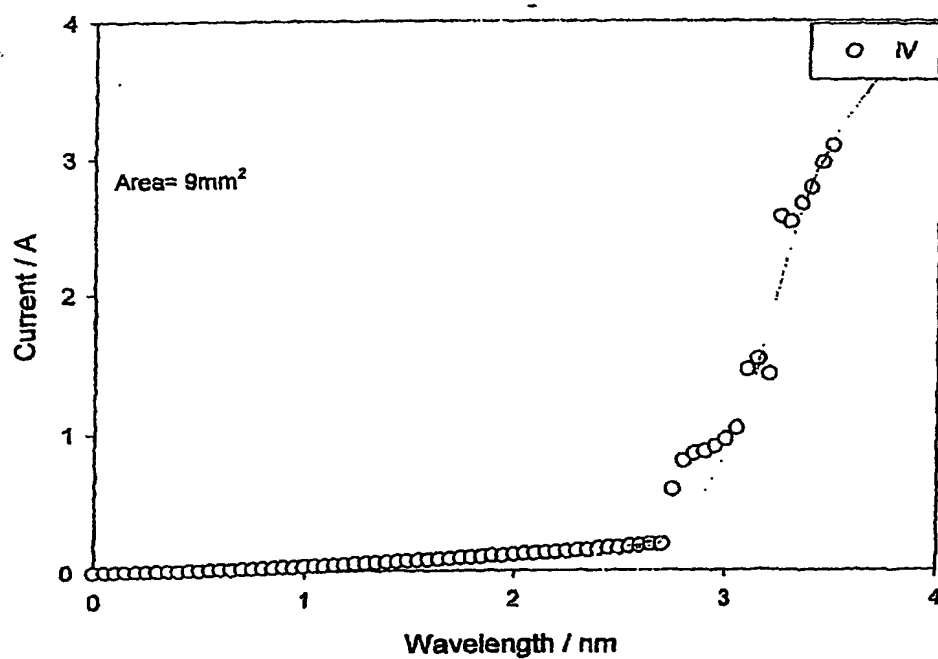


Fig. 16

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